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Thermal/hyperthermal Collision Dynamics of Atmospheric Species

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14. ABSTRACT

Shot-noise limited, direct absorption IR laser methods have been used to study (i) state-to-state reaction dynamics in crossed supersonic jets and (ii) temperature dependent studies of OH + O_3 chemical chain reaction kinetics via flash kinetic spectroscopy. In this first area, state-resolved studies of F + H_2 scattering have been completed from E_{coll} = 2.4 kcals/mole down to 0.30 kcals/mole, which by virtue of the high quantum state resolution in product detection have revealed contributions due to *non-adiabatic* reactions with spin orbit excited F* atoms. These methods have been extended to F + CH_4, where high resolution IR laser Dopplerimetry can be used to extract product state velocities and thus differential cross section information at the state-to-state level. Time-resolved flash kinetic studies of the OH/HO_2/O_3 chemical chain reaction has been performed from 300 K down to 190 K, providing first access to temperature conditions relevant to accurate modeling of the lower stratosphere. These methods have been extended to study "airglow" dynamics of highly rotationally excited OH(v,N) radicals formed from H + O_3 reactions.

15. SUBJECT TERMS

state-to-state collision dynamics, atmospheric radical kinetics

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Equipment supplement to F49620-97-1-0038
Final report:

The funds have been used to purchase and install new equipment for significantly expanding the current capabilities of both the i) high resolution photolysis spectrometer for study radical kinetics and the ii) crossed supersonic jet system for study of state-resolved reaction dynamics. These upgrades include a Kr+ laser, excimer laser, optical parametric oscillator, pulsed dye laser, non-linear frequency mixing crystals, as well as electronics for REMPI/LIF detection. We are now in the process of converting the current IR based detection of final product states to the less general albeit much more sensitive LIF/REMPI methods. Our first test project with the upgraded system will be to use the new OPO light source for selective excimer photolysis dynamics of X-H stretch excited molecules from the v=2 manifold. The next stage will involve looking at reaction dynamics of vibrationally excited species with radicals formed in the discharge supersonic expansion sources developed under this grant. A later stage will involve looking at state-to-state reaction dynamics of hypersonically excited reagents using both translational and vibrational excitation.

Papers published, in press or submitted under the current granting period acknowledging AFOSR support

1) S. A. Nizkorodov, W. W. Harper, B. W. Blackmon and D. J. Nesbitt, "Temperature dependent kinetic studies of the $OH/HO_2/O_3$ chain reaction by time resolved high resolution laser absorption spectroscopy", J. Phys. Chem (in press).

2) W. W. Harper, S. A. Nizkorodov, and D. J. Nesbitt, "Quantum state-resolved reactive scattering of $F + CH_4 \rightarrow HF(v,J) + CH_3$: Nascent HF(v,J) product state distributions", J. Chem. Phys. (submitted).

Invited talks during the current AFOSR granting period

"Single particle microscopy above and below the diffraction limit", Optical Society of America, Santa Clara, CA, September 28, 1999.

"Chemical physics with lasers: From slit jet discharges to single molecule spectroscopy", Department of Chemistry, University of Wisconsin, Madison, WI, October 26, 1999.

"Where Chemistry meets Physics", CU Wizards Science Outreach Program, Department of Chemistry, University Colorado, Boulder, CO, October 30, 1999.

"From state-to-state reaction dynamics to single molecule microscopy", Department of Chemistry, University of Maryland, College Park, MD, November 11, 1999.

"Chemical dynamics with a twist: From state-resolved reactions in supersonic jets to single molecule microscopy", Department of Chemistry, University of Southern California, Los Angeles, CA, January 10, 2000.

"Chemical kinetics with a twist: From state-to-state reaction dynamics to single molecule microscopy", Department of Chemistry, University of Arizona, Tuscson, AZ, January 24, 2000.

"Microscopy at and below the diffraction limit via resonant scattering and laser induced fluorescence: Recent progress from apertureless NSOM", American Physical Society, Minneapolis, MN, March 21, 2000.

"Probing quantum state to state dynamics: From clusters to chemical reactions", American Chemical Society (219th national Meeting), San Francisco, CA, March 26, 2000.

"From Single Collisions to Single Molecules", Institute for Physical Chemistry, University of Goettingen, Goettingen, Germany, April 13, 2000

"Spectroscopy above and below the diffraction limit", Max Planck Institute for Biophysical Chemistry, Goettingen, Germany, April 28, 2000.